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# NANOPARTICULATE FLUX PINNING CENTERS FOR $YBa_2Cu_3O_{7-\delta}$ FILMS (POSTPRINT)

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#### 14. ABSTRACT

 $YBa_2Cu_3O_{7-\delta}$  high temperature superconductors can maintain fairly high critical current densities ( $J_c$ ) with increasing magnetic field. This in-field performance can be further improved upon by incorporating nanoparticulate magnetic flux pinning centers into the superconductors. This short paper briefly discusses and compares recent efforts by the U.S. Air Force Research Laboratory to incorporate insulating nanoparticles into the YBCO superconducting thin films by pulsed laser deposition.

#### 15. SUBJECT TERMS

high temperature, superconductors, in-field, nanoparticulate, magnetic, flux pinning, critical current densities, thin films

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# Nanoparticulate Flux Pinning Centers for $YBa_2Cu_3O_{7-\delta}$ Films

Paul N. Barnes, Joseph W. Kell, Brandon C. Harrison, Timothy J. Haugan, Jack L. Burke, and Chakrapani V. Varanasi

 $Abstract{-}YBa_2Cu_3O_{7-\delta}$  high temperature superconductors can maintain fairly high critical current densities  $(J_{\rm c})$  with increasing magnetic field. This in-field performance can be further improved upon by incorporating nanoparticulate magnetic flux pinning centers into the superconductors. This short paper briefly discusses and compares recent efforts by the U.S. Air Force Research Laboratory to incorporate insulating nanoparticles into the YBCO superconducting thin films by pulsed laser deposition.

*Index Terms*—Flux pinning, high-temperature superconductors (HTS), rare earth doping, YBCO.

#### I. INTRODUCTION

The pinning properties of  $YBa_2Cu_3O_{7-\delta}$  (YBCO) make this superconductor desirable for use in applications in the form of biaxally aligned YBCO coatings on buffered metallic substrates [1]. The Jc can be greatly increased in the YBCO films by the intentional addition of a high density of nanoparticles into the superconductor itself. Nanoparticulate dispersions have been accomplished by a variety of methods [2]–[6]. For maximum benefit as 3-D pinning centers, the nanoparticulates must be well dispersed and occupy less than 15% of the superconductor's volume. To minimize cooling requirements, emphasis is generally on the 60 K–77 K operating range for many applications [1].

Pinning methods discussed below include recent efforts by the U.S. Air Force Research Laboratory to improve the pinning properties of the YBCO via pulsed laser deposition (PLD). Table I summarizes the particular pinning methods being considered here. Pinning by the inclusion of Y<sub>2</sub>BaCuO<sub>5</sub> (Y211) phase nanoparticulates in a multilayered deposition process has previously undergone some optimization. However, pinning by a pie wedge sector in the PLD target (non-layered nanoparticulate dispersion) and minute doping with deleterious rare earths have been more recently developed and have undergone little optimization up to this point. Relative performance of these pinning materials and the processes will likely change as the

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TABLE I
LISTING OF THE PINNING PROCESSES COMPARED

[Ref.]	Process for Incorporating Pinning Centers in YBCO	Pinning Material
[6]	minute rare earth doping (% of Y substitution)	Tb (1%) Ce (0.1%) Nd (0.1%)
[5]	non-layered nanoparticulate dispersion (dual-sector PLD target)	$\begin{array}{l} BaSnO_3 \\ Y_2BaCuO_5 \end{array}$
[3]	multilayered nanoparticulate dispersion (alternating PLD targets)	Y <sub>2</sub> BaCuO <sub>5</sub>

Results presented may not necessarily be thoroughly optimized, although are the latest reported results.

methods become more fully developed. Future optimization will also help to distinguish between the benefit of the pinning material and the particular process for incorporating the pinning centers.

#### II. PINNING METHODS

#### A. Minute Doping

It was recently demonstrated that adding very minor amounts,  $\leq 1\%$  substitution of Y, of the typically deleterious rare earths (RE) into quality YBCO can provide substantial pinning [6]. The advantage of this minute doping is that the pinning enhancement can be achieved using these dopants in YBCO thin films processed under the same conditions as the plain YBCO films. By removing the necessity of individualized optimization, YBCO coated conductors can be more readily tailored for a desired performance.

The divalent rare earth elements have been noted in the literature as not readily forming the proper REBCO phase in bulk form [7]. When used in quantities that other REs enhance performance, typically  $\geq \! 10\%$  Y substitution, they degrade the YBCO quality. Even if a particular rare earth, such as Nd and La, can readily form the proper REBCO superconducting phase, they can substitute undesirably into the Ba site degrading the superconductor's performance.

Since these undesirable dopants are degrading in nature, when used in minute quantities as  $(Y_{1-x},RE_x)Ba_2Cu_3O_{7-\delta}$  where  $x\leq 0.01$ , or  $\leq 1\%$ , the proper density of defects may be established. Such small quantities will require that the RE dopant is sufficiently dispersed. Even so, it is not certain as to whether the dopants primarily result in site substitution somewhere in the lattice or form secondary phase inclusions, or potentially both.

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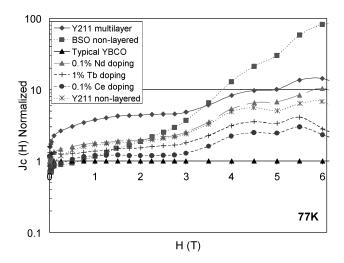


Fig. 1.  $J_c(H)$  curves for pinned YBCO normalized to the  $J_c(H)$  curve for YBCO at 77 K. The slight "wave" of the curves at higher fields is due to the minor increase in the sample measurement error and choice of the particular point. This is for H/c-axis.

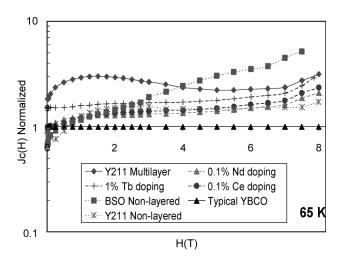


Fig. 2.  $J_c(H)$  curves for pinned YBCO normalized to the  $J_c(H)$  curve for YBCO at 65 K. The slight "wave" of the curves at higher fields is due to the minor increase in the sample measurement error and choice of the particular point. This is for H//c-axis.

Fig. 1 provides the pinning information where the  $J_c(H)$  curves are normalized with respect to the  $J_c(H)$  curve for a typical YBCO sample. At 77 K, the best pinning improvement in YBCO among the tested dopants for the initially chosen doping levels was the 0.1% Nd for Y. This is likely a result of the Nd substituting into Ba sites in the unit cell generating the pinning centers. At 65 K, the best enhancement occurred for a 1% Tb for Y doping. Fig. 2 provides the pinning information at 65 K where the  $J_c(H)$  curves are normalized with respect to the  $J_c(H)$  curve for YBCO.

Initial concentrations attempted were restricted to 10%, 1%, 0.1% and 0.01%. In general, for the dopants studied, replacing Y with about 0.1% dopant tended to improve the infield properties of the resulting YBCO across both temperature regimes with the exception of 1% Tb.

Optimization of the doping levels between these values will likely show further improvements and lead to varying levels of

the different dopants for peak performance. A question that remains is whether the minute dopants are actually site substituting with the Y or causing secondary phase precipitates. There are initial indications that nanoparticulates are being formed by the dopants.

#### B. BSO Non-Layered Nanoparticulate Dispersions

In this approach, a special YBCO PLD target was fabricated with a pie sector inclusion of BaSnO<sub>3</sub> (BSO) [5]. As the target was rotated, both YBCO and BSO ablation occurred allowing BSO nanoparticles to be incorporated into the YBCO. With high lattice mismatching and potential reactivity, the BSO nanoparticles may provide different pinning than other inclusions. Comparing the dual-sector target approach for non-layered pinning with Y211 will be instructive after additional optimization has occurred. In the future, this may help to distinguish between the chosen pinning process being used and the selected material for pinning with respect to the effect on pinning performance.

Magnetization data obtained shows a significant increase in the  $J_{\rm c}$  of the BSO pinned YBCO films at higher fields. Refer to Fig. 1 and Fig. 2. This occurred at both 77 K and 65 K compared to the typical in-field  $J_{\rm c}$  for YBCO. An order of magnitude increase in  $J_{\rm c}$  was achieved at 4 T, 77 K. The decrease in  $J_{\rm c}$  at low fields may be inherent to the BSO pinning or simply that further optimization of process is necessary. A similar decrease in the self-field  $J_{\rm c}$  was also observed in SmBCO films with low  $T_{\rm c}$  nanoparticles even though a higher  $J_{\rm c}$  was observed at high fields [8].

Another important effect of the BSO not seen in the figures is that  $H_{\rm irr}$  was significantly increased for the BSO pinned YBCO films as compared to plain YBCO. The  $H_{\rm irr}$  was  ${\sim}8.5$  T at 77 K and  ${\sim}13.4$  T at 65 K. Scanning electron microscopy of the surface verified that a dense distribution of nanoparticles existed throughout the films. The nanoparticles were  ${\sim}10$  nm in size. Additional cross-sectional TEM is being performed but past samples of Y211 pinned YBCO using the dual-sector PLD target for pinning demonstrated the presence of the nanoparticulate dispersion throughout the film. Non-layered Y211 pinned YBCO is also displayed in the figures.

#### C. Pinning Comparison

Discussion of the multilayered Y211 pinned YBCO is not given here, but has been presented previously [3]. Pinning by this approach is also displayed in Fig. 1 and Fig. 2. At high fields, the BSO pinned samples clearly provide the highest pinning. Of interest is the peaking effect of the multilayered Y211 pinned YBCO at just over 1 T. This may well be due to the influence of the particular spacing of the layers in the film; even so, the layered structure will likely have a greater effect on current values when the magnetic field is oriented in the ab-plane. The increase of all the curves at higher fields is indicative of a higher  $H_{\rm C2}$  than typical YBCO for the various samples. This is especially noteworthy in the BSO-pinned YBCO films.

#### III. CONCLUSION

Pinning of superconducting YBCO is demonstrated by a variety of methods and materials. Each pinning method and each material has its own unique advantages and pinning properties.

At the present, the advantages of the three magnetic flux pinning methods compared in this paper are: (1) the Y211 multilayered pinning provides the best lower field, <3–4 T, improvements in the current density, (2) the BSO pinning provides that best high field pinning and extension of the irreversibility field to higher levels, and (3) the minute doping is the simplest, most general processing method for providing some overall pinning enhancement. Further optimization of these pinning strategies will result in not only greater improvements to the YBCO in-filed critical current densities but may provide different functional relationships to each other as well.

#### REFERENCES

[1] P. N. Barnes, M. D. Sumption, and G. L. Rhoads, *Cryogenics*, vol. 45, pp. 670–686, 2005.

- [2] S. Kang, A. Goyal, J. Li, A. A. Gapud, P. M. Martin, L. B. Heatherly, J. R. Thompson, D. K. Christen, F. A. List, M. Paranthaman, and D. F. Lee, *Science*, vol. 311, pp. 1911–1914, 2006.
- [3] T. J. Haugan, P. N. Barnes, R. Wheeler, F. Meisenkothen, and M. Sumption, *Nature*, vol. 430, pp. 867–870, 2004.
- [4] J. Hanisch, C. Cai, R. Huhne, L. Schultz, and B. Holzapfel, *Appl. Phys. Lett.*, vol. 86, p. 122508, 2005.
- [5] C. V. Varanasi, P. N. Barnes, J. Burke, J. Carpenter, and T. J. Haugan, Appl. Phys. Lett., vol. 87, p. 262510, 2005.
- [6] P. N. Barnes, J. W. Kell, B. C. Harrison, T. J. Haugan, C. V. Varanasi, M. Rane, and F. Ramos, *Appl. Phys. Lett.*, vol. 89, p. 012503, 2006.
- [7] T. J. Haugan, J. C. Tolliver, J. M. Evans, and J. W. Kell, "Crystal chemical substitutions of Yba<sub>2</sub>Cu<sub>3</sub>O<sub>7−6</sub> to enhance flux pinning," in HTS Thin Film and More on Vortex Studies, A. Narlikar, Ed. New York: Nova Science Publishers Inc., 2005, vol. 49.
- [8] M. Miura, Y. Yoshida, Y. Ichino, Y. Takai, K. Matsumato, A. Ichinose, S. Horii, and M. Mukaida, *Jap. J. Appl. Phys.*, vol. 45, pp. L11–L13, 2006.